PHYTOREMEDIATION OF SOIL CONTAMINATED BY LEAD AND COPPER IN FARNHAM, QUEBEC

GREENHOUSE TESTS AND TECHNO-ECONOMIC EVALUATION

FINAL REPORT

(Ref no.: EC9045)

PRESENTED TO

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May 2000

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May 2000

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ABSTRACT

The purpose of this project was to evaluate the technical and economic feasibility of phytoremediation to reclaim soil contaminated with lead and copper in a confining cell at the Canadian Forces base in Farnham, in the Eastern Townships. Specifically, the goal was to find an ecological way of reducing lead and copper content of the upper soil layer (0.0 to 0.8 m, that is, about 1556 m^3 of soil) to meet level C criteria of the Environnement Québec (MENV) «*Politique de protection des sols et de réhabilitation des terrains contaminés*»—less than 1000 mg/kg for lead and less than 500 mg/kg for copper.

Phytoremediation is a biological technique using vascular plants (trees, shrubs or herbaceous plants) to remove, immobilize or degrade organic or inorganic contaminants in soil, water or sediment.

Specific objectives of the project were as follows:

1. To conduct a test using input that can promote optimal growth of plants able to accumulate metal in their tissue. To this end, the following were evaluated:
   - Two chelating agents, citric acid and ethylene diamine tetraacetic acid (EDTA), for removal of lead and copper from the soil
   - Two plants that can accumulate metal and generate a large aerial biomass: Indian mustard (*Brassica juncea*) and corn (*Zea mays*)
   - Addition of fertilizers (nitrogen, phosphorus and potassium) to promote optimal growth of plants in the sandy loam

2. To evaluate the technical feasibility of phytoremediation of soil in the Farnham confining cell based on results of greenhouse tests. The goal was to lower lead content (initially 1200 mg/kg) to meet the level C criterion of the MENV policy; at 165 mg/kg, the initial copper content was already below the level C criterion.

3. To estimate the costs of phytoremediation and the decontamination period required to meet the level C criterion of the MENV policy.

The initial soil analysis showed that the lead content had to be lowered by at least 200 mg/kg to meet the decontamination objectives. Best results were obtained with *Brassica juncea* plus EDTA (3.0 g/kg of soil) as chelating agent, which yielded a lead extraction rate of 5.55 mg/kg of soil. At this rate, 36 harvests would be required to meet project objectives; that is, it would take 18 years, since two harvests a year are possible with mustard.

The EDTA alone costs $25 per tonne of soil to be treated, and it must be applied for each harvest. In other words, phytoremediation by itself does not seem to be a realistic

option for remediation of the Farnham soil. It could, however, be considered in combination with some other technology; given that the soil is confined in a watertight facility, for example, the leachate and the metals solubilized in it could be recovered.

Costs are very high and treatment time very long compared to other methods of remediation—two major factors against phytoremediation compared to standard technologies used (such as secure landfill) that prevent its recommendation as sole method of decontaminating the soil in the Farnham facility.

RÉSUMÉ

Ce projet visait à évaluer la faisabilité technico-économique de la phytorestauration pour réhabiliter des sols contaminés au plomb et au cuivre, entreposés dans une cellule sur le terrain de la Garnison de l'Estrie des Forces canadiennes à Farnham. Plus précisément, l'objectif général consistait à trouver un moyen écologique pour ramener la concentration du plomb et du cuivre de la couche superficielle de sol (0,0 à 0,8 m, c'est à dire environ 1 556 m^3 de sol), sous le critère «C» de la «*Politique de protection des sols et de réhabilitation des terrains contaminés*» du ministère de l'Environnement (MENV) (ci-après appelée «la Politique» : 1 000 mg/kg pour le plomb et 500 mg/kg pour le cuivre).

La phytorestauration est une technique biologique qui utilise les plantes vasculaires (arbres, arbustes ou plantes herbacées) pour réhabiliter, stabiliser ou extraire les contaminants organiques ou inorganiques dans le sol, l'eau ou les sédiments.

Les objectifs spécifiques au projet à l'étude étaient les suivants :

1. faire un essai avec des intrants susceptibles de favoriser la croissance optimale de plantes pouvant accumuler des métaux dans leurs tissus. Pour ce faire, les paramètres suivants ont été évalués :
   - 2 agents chélateurs, soit l'acide citrique et le «*Ethylene Diamine Tetraacetic Acid*» (EDTA), en vue d'extraire le plomb et le cuivre du sol;
   - 2 plantes pouvant accumuler des métaux et produire une biomasse aérienne importante. Il s'agit de la moutarde indienne (*Brassica juncea*) et du maï s (*Zea mays*);
   - ajout de fertilisants (azote, phosphore et potassium) dans le but de favoriser la croissance optimale des plantes dans le sol (loam sableux).

2. évaluer la faisabilité technique de la phytorestauration du sol confiné dans la cellule de Farnham à partir des résultats obtenus lors d'essais en serre. Il s'agissait de réduire les teneurs en plomb (1 200 mg/kg initialement) sous le

critère «C» de la Politique du MENV, le cuivre (165 mg/kg initialement) se situant déjà sous ce critère;

3. estimer les coûts de la phytorestauration et de la durée de la décontamination permettant d'atteindre le critère «C» de la Politique.

Selon les résultats initiaux des analyses de sol, il faut abaisser d'au moins 200 mg/kg de sol la teneur en plomb pour atteindre l'objectif de décontamination pour ce métal. À la lumière des résultats obtenus, le meilleur rendement a été obtenu avec *Brassica juncea* et le EDTA (3,0 g/kg de sol) comme agent chélateur. Le taux d'extraction du plomb correspond dans ce cas à 5,55 mg/kg de sol. Il faudrait donc 36 récoltes pour atteindre les objectifs visés, travaux qui s'étendraient alors sur 18 années, puisqu'il semble possible d'obtenir annuellement 2 récoltes avec la moutarde.

Le coût du EDTA à lui seul implique un déboursé de 25 $ la tonne de sol à traiter, et son application doit être répétée à chaque récolte. La phytorestauration ne semble donc pas constituer, à elle seule, une avenue réaliste pour la réhabilitation des sols de Farnham. Il pourrait être envisageable de combiner cette technologie avec une autre, par exemple en tirant profit du fait que les sols sont confinés dans une cellule étanche et en récupérant le lixiviat et les métaux qui y seraient solubilisés.

Les coûts et le temps de traitement très élevés comparativement à d'autres moyens de réhabilitation défavorisent grandement la phytorestauration par rapport aux autres technologies usuelles telle que la mise en cellule sécuritaire et empêchent de la recommander comme approche unique pour décontaminer les sols de la cellule de Farnham.

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1 INTRODUCTION

Former use of cartridges with lead and copper bullets for target practice at the Canadian Forces firing range in Farnham in the Eastern Townships led to lead and copper contamination of the soil in the stop butts, which was eventually placed in a confining cell. The Department of National Defence launched a phytoremediation research project with the goal of reducing the lead and copper content of the contaminated soil. The project described herein was carried out as part of a scientific cooperation agreement signed in August 1998 between Environment Canada, Laval University and Biogénie S.R.D.C. Inc. (Biogénie).

The following people worked on this project:

- Didier Barré and Daniel Bradshaw, Defence Construction Canada
- Dr. Antoine Karam, Laval University
- Lucie Olivier, Environment Canada
- Frédéric Schooner, Environment Canada
- Yvan Pouliot, Biogénie
- Nicolas Moreau, Biogénie
- Gervais Lessard, Biogénie

2 BACKGROUND AND PROBLEM

2.1 CONTAMINATION HISTORY

The old practice range where the contaminated soil originated is located in the middle of the Eastern Townships base in Farnham. The range was used for military training from the beginning of the 1940s to the end of 1996, particularly for rifle shooting at fixed targets. Over the years, the soil of the stop butts became contaminated by projectile fragments of lead and copper.

According to the description in a report by the firm Sanexen (1998), the surface of the stop butts was reworked several times with bulldozers to slow erosion. In addition, layers of fill were added over the years.

2.2 CHARACTERIZATION

Before the excavation and confinement performed by Sanexen in 1998, three site characterization studies were conducted. The analyses performed during these studies were used to classify the copper and lead contamination of the soil according to the generic criteria of the 1999 Environnement Québec «*Politique de protection des sols et de réhabilitation des terrains contaminés*» (MENV policy). Table 1 shows the MENV policy criteria.

Table I: Generic Soil Criteria

Metal	Soil Criteria (mg/kg dry matter)		
	Level A[1]	Level B[2]	Level C[3]
Lead	50	500	1000
Copper	40	100	500

1. Background concentrations for inorganic parameters and quantification limit for organic parameters. The quantification limit is the minimum concentration that can be quantified with a given method of analysis with a specified reliability.

2. Maximum acceptable limit for lands used for residential, recreational or institutional purposes or lands used for commercial purposes in a residential area. Institutional purposes include use of the land for hospitals, schools and daycare centres. Recreational purposes cover a wide variety of uses, some of which are more sensitive than others. Hence in the case of sensitive uses, such as a playing field, content must meet level B criteria; for recreational uses that are considered less sensitive, such as bicycle paths, level C criteria are satisfactory.

3. Maximum acceptable limit for commercial lands in non-residential areas and for industrial sites.

The study conducted by Groupe Cartier in 1995 examined only four soil samples from the stop butts to a depth of 0.6 m. Analysis results showed lead and copper concentrations were below level B criteria. However, visual observations indicated presence of numerous projectile fragments in the sandy soil of the stop butts.

In 1996, SM Environnement took 46 core samples from the two stop butts using manual soil augers. The core samples ran to an average depth of 1.1 m, ranging from 0.5 to 4.1 m. Analysis of these samples showed that lead and copper contamination of the stop butts exceeded level B criteria of the MENV policy to a depth of 2.2 m.

A second study by SM Environnement, conducted in 1998 using samples from 26 test trenches, showed that the soil contained a large quantity of bullet fragments. Sieve tests showed that most of these fragments measured 5 to 10 mm. It was estimated that about 1850 m^3 of soil did not meet level C criteria for copper and lead.

2.3 PLACEMENT OF CONTAMINATED SOIL IN THE CONFINING CELL

In 1998, Sanexen excavated the soil, built the confining cell and placed the segregated contaminated soil in it. A total of 4790 m^3 of soil contaminated by lead and copper were processed; the soil was run through 5-mm and 10-mm sieves and the fraction between 5 and 10 mm (which included the largest proportion of bullet fragments) was placed in the Cintec landfill site in Ville Lasalle.

The rest of the soil was as follows:

- 562 m^3 (12.7%) had lead and copper concentrations below level B criteria
- 2302 m^3 (52.0%) had lead and copper concentrations between level B and level C criteria
- 1556 m^3 (35.2%) had lead and copper concentrations higher than level C criteria

The soil with lead and copper concentrations between level B and level C criteria was placed in the bottom of the cell, covering it almost entirely in a layer 1.5 m thick. Soil with a grain size of more than 10 mm, containing some bullet fragments, was placed at the west end of the confining cell. The most contaminated soil (exceeding level C criteria) was placed in a layer 0.3 to 0.8 m thick in the upper part of the cell and covered with an impermeable membrane. The confining cell is 97 m long and 21 m wide.

2.4 PROBLEM

The presence of the confining cell at the base restricts armed forces training activities. A decision was made, accordingly, to reduce the lead and copper content of the soil in the cell, so the reclassified soil can then be disposed of more easily and the cell dismantled.

3 PURPOSE AND SPECIFIC OBJECTIVES

The purpose of this project was to determine the technical and economic feasibility of using phytoremediation to decontaminate the soil contaminated by lead and copper in the Farnham confining cell (97 m long, 21 m wide and 1.5 m deep). Specifically, the goal was to find an ecological way of reducing the lead and copper content of the upper soil layer (0.0 to 0.8 m) to meet level C criteria of the MENV policy.

3.1 PHYTOREMEDIATION

Today, sites contaminated by heavy metals are generally rehabilitated using physicochemical technologies: chemical treatment, leaching, vitrification, thermal treatment, electrokinetic treatment, dig and dump, etc.

Phytoremediation, a biological process, is an exception to this rule. Phytoremediation is the use of vascular plants (trees, shrubs and herbaceous plants) to remove, degrade or immobilize organic or inorganic contaminants in soil, water or sediment.

Depending on the type of contaminant, the environment affected and the method used by the plant to reduce or control contamination, phytoremediation falls into six categories: phytoaccumulation, phytodegradation, phytostabilization, phytovolatilization, rhizodegradation and rhizofiltration (ITRC, 1999).

Phytoaccumulation, also called phytoextraction, involves uptake of metal contaminants in the soil by the roots of the plant and their transport to aerial tissue. Some plants are hyperaccumulators, that is, they are capable of absorbing very large quantities of metal compared to other plants and to background concentrations in the soil. These plants are selected and planted on sites that require rehabilitation based on the metals present and other site conditions. When the plants have finished growing (this may take a few weeks or a few months), the plants are harvested; the plant biomass and the metals extracted from the soil can then be disposed of by landfill, incineration or composting. The plants may be seeded and harvested repeatedly, if necessary, to meet decontamination objectives. Analysis of the plant tissue harvested (stems, leaves, roots, etc.) determines their final destination or use.

Phytoremediation performance depends on availability of the metals to the plants. And since the main characteristic of hyperaccumulators is their capacity to synthesize substances that can solubilize metals in the rhizophere, the possibility of artificially reproducing these conditions was considered for this study. Studies of lead phytoextraction (Huang *et al.*, 1997 a and b) clearly demonstrate that artificial chelating agents such as ethylene diamine tetraacetic acid (EDTA) can increase lead content in the plant tissue from less than 500 mg/kg to more than 10 000 mg/kg.

Soil pH is also a key factor because it controls the solubility of metals in the soil. It has also been demonstrated that reducing pH reduces adsorption of metals to soil particles, which increases their solubility and their availability to the plants.

3.2 REMEDIATION METHODOLOGY

As with other site rehabilitation technologies, an in-depth study is required before beginning phytoremediation of a metal-contaminated site. The design of the phytoremediation system depends on contaminants present, site conditions, decontamination objectives, plants used and input required for the success of the project. In general, the following are key steps in the remediation process:

- ***Characterization***

Characterization is performed to identify contaminants and determine their concentrations. It also shows the extent and depth of contamination.

- ***Plant selection***

A species or several species are selected based on contaminant or contaminants present and physical characteristics of the soil to be treated.

- ***Treatability tests***

Treatability tests, generally conducted in a greenhouse, are performed to determine in advance whether objectives can be met. They are also used to evaluate total duration of the treatment.

- ***Input (irrigation, fertilizers, chelating agents)***

Soil analyses performed prior to the greenhouse tests are used to determine fertilizers and irrigation required and to select chelating agents to solubilize contaminants.

- ***Collection of groundwater***

Groundwater collection is considered where irrigation or application of chelating agents during the treatment might cause contaminants to migrate to the groundwater. Water is pumped downstream of the contaminated site and recirculated into the irrigation system, or an impermeable membrane is installed under the soil to be rehabilitated.

- ***Measurement of decontamination rate***

The rate of decontamination is measured during greenhouse tests as well as during the first season in the field. These measurements are used to determine how long the remediation will take.

➢ ***Site preparation and seeding***

The land is prepared as land is generally prepared for crops; the same machinery and methods are used.

➢ ***Monitoring***

Growth of the plants must be monitored to ensure rapid response to nutritional deficiencies, insect attack or disease. If necessary, the plants must be irrigated.

➢ ***Harvest***

Whenever possible, the entire plant is harvested (roots and aerial parts). However, with most species used, only the aerial part is harvested.

➢ ***Management of harvested plants***

The plants harvested must be analyzed to determine contaminant content and hence the method of management. If the phytoremediation process has worked well, the metal content of the harvested plants is generally high enough for them to be considered hazardous material. To minimize disposal costs, plant mass and volume must be reduced by drying or incineration.

3.3 SPECIFIC OBJECTIVES OF THE FARNHAM PROJECT

The specific objectives of the Farnham project were as follows:

1. To conduct a test using input that can promote optimal growth of plants able to accumulate metal in their tissue. To this end, the following were evaluated:
   - Two chelating agents, citric acid and ethylene diamine tetraacetic acid (EDTA), for removal of lead and copper from the soil
   - Two plants that can accumulate metal and generate a large aerial biomass: Indian mustard and corn
   - Addition of fertilizers (nitrogen, phosphorus and potassium) and an organic amendment to promote optimal growth of plants in the sandy soil

2. To evaluate the technical feasibility of phytoremediation of the soil in the Farnham cell based on results of greenhouse tests. A project schedule must then be developed for decontamination of the entire cell to lower lead content (initially 1200 mg/kg) to meet the level C criterion of the MENV Policy; at 165 mg/kg, the initial copper content was already below the level C criterion.

3. To estimate the costs of phytoremediation and the decontamination period required to meet the level C criterion of the MENV policy.

4 PROTOCOL FOR PHYTOREMEDIATION GREENHOUSE TESTS

The protocol selected for the phytoremediation tests for this project comprises the following:

- Two plant species: Indian mustard (*Brassica juncea*) and corn (*Zea mays*), silage variety 4066
- Chelating agent: citric acid (2 doses: 0.8 and 1.6 g/kg of soil), a negative control (no chelate added) and a positive control (EDTA: 3.0 g/kg of soil)
- Plant culture in 22-L pails, six plants per pail for mustard and two plants per pail for corn
- Duplicate tests for the mustard and triplicate tests for the corn

Twenty pails were required for the tests *(1 species x 4 chelates (2 doses + 2 controls) x 2 duplicates and 1 species x 4 chelates x 3 triplicates)*, for a total of 48 Indian mustard plants and 24 corn plants.

The species were selected based on their known metal accumulation performance and their large biomass (McIntyre, 1999). A literature review was conducted as well as tests in soil from Île-aux-Corbeaux (Olivier *et al,* 1999) before the choice was made. A search for perennials with known capacity for accumulating heavy metals proved fruitless;[1] with a perennial, seeding and harvesting operations would be minimal, and hence costs would be much lower.

Dr. Antoine Karam suggested that citric acid, an organic acid, be used during the metal phytoextraction as a solubilization agent, to make the metals available to the plants before they are harvested. Though citric acid forms metal chelates which are generally not as stable as EDTA-metal chelates, it may be a more economical option than EDTA.

The soil samples taken at Farnham were analyzed for the following: available and total nitrogen (N), available phosphate (P), potassium (K), lead (Pb), copper (Cu), pH and total organic carbon (TOC). Table II shows the results. Since the soil collected contained only 0.22% TOC (0.34% organic matter), it was amended with compost to increase the organic matter content to about 3% (weight/weight). Visual observation suggests the soil can be described as sandy. Chemical fertilizer (nitrogen, phosphorus and potassium) was also applied, as per the recommendations for corn in the fertilization guide of the Quebec Fertilizer Manufacturers Association.

1. Some 1500 scientific publications in the field of phytoremediation were reviewed.

Table II: Soil Analysis Results

	Lead (mg/kg)	Copper (mg/kg)	N* (mg/kg)	P** (mg/kg)	K (mg/kg)	TOC*** %C	pH
Initial							
Sample 1	1 000	150					6.1
Sample 2	1 200	180					6.0
Average	1 100	165	10.3	180	177	0.22	6.1
Final							
Corn (control)	1 300	260					5.8
Corn (EDTA), top	280	80					7.0
Corn (EDTA), bottom	640	120	<3	150			
Corn (Citric acid, X1)	800	120					5.3
Corn (Citric acid, X2)	760	130					5.4
Mustard (control)	1 000	170					6.4
Mustard (EDTA), top	360	100					8.0
Mustard (EDTA), bottom	1 200	150	9	210			
Mustard (Citric acid, X1)	880	140					5.8
Mustard (Citric acid, X2)	960	140					5.2

* Available soluble nitrogen
** Available phosphate
*** Total organic carbon

Note: The initial analyses of nitrogen, phosphorus, potassium and TOC concentrations were made on soil sampled on September 21, 2000.

The plants were grown under sodium lamps, using a cycle of 14 hours of light and 10 hours of darkness, and they were watered as needed (four to five times a week).

The Indian mustard was harvested in mid-January 2000, after nine weeks of growth, and the corn at the end of January 2000, after 11 weeks of growth. The aerial parts of the plants were weighed (to determine dry weight) and analyzed for lead and copper content, to determine the quantities of these metals extracted from the soil. The mustard roots were also harvested and then weighed and analyzed for lead and copper content. The corn roots, which are very hard to remove from the soil, were not harvested.

In all cases, a germination test was performed after the harvest to check that the medium was not rendered toxic by the addition of a chelating agent. All plants were cut into pieces of about 5 cm and dried at 80°C for 72 hours. All samples were weighed and then pulverized to 2 mm (crushed and then passed through a sieve) before being analyzed.

5 ACTIVITIES

5.1 CHRONOLOGY

Table III summarizes the activities conducted to date. Note that the soil collection that took place on September 21, 1999, had to be repeated because lead and copper concentrations measured in the soil collected were below level C of the MENV policy. Since the purpose of the project was to lower lead and copper concentrations to meet criteria C, the tests had to be performed on soil contaminated beyond this level. A second set of twenty 22-L pails of soil was collected on October 27, 1999. Table II shows the results of the analysis of this soil.

One dose of EDTA (3 g/kg of soil) and two doses of citric acid (0.8 and 1.6 g/kg of soil) were added to both species. Both citric acid doses were divided in two, with the first half applied two weeks before harvesting and the second one week before.

Table III: Greenhouse Testing Activities

Date	Activity	Comments
September 8, 1999	Startup meeting	Present: A. Karam, L. Olivier, F. Schooner, N. Moreau, G. Lessard
September 20, 1999	Greenhouse reservation	Laval University greenhouses (to January 2000)
September 21, 1999	Site visit (Farnham) and soil collection	Present: A. Karam, L. Olivier, F. Schooner, D. Bradshaw, G. Lessard. Twenty 20-L pails of soil collected
October 12, 1999	Drafting of greenhouse testing protocol	
October 27, 1999	Additional soil collection at Farnham	Soil collection had to be repeated because contamination level of the soil collected on September 21 was too low for the needs of the study
November 1, 1999	Soil preparation	Application of fertilizers and organic amendment
November 12, 1999	Seeding	Corn: four seeds per pail Mustard: 10 to 12 seeds per pail
November 26, 1999	Corn plants placed under lights	2 plants per pail
December 14, 1999	Greenhouse visit Mustard plants placed under lights	Present: A. Karam, L. Olivier, Y. Pouliot, G. Lessard 6 plants per pail
January 4, 2000	Plants removed from tables Second application of fertilizer to corn plants	Plants are too tall and could be burned by the sodium lamps. The pails are placed directly on the ground.
January 5, 2000	Application of first half of both doses of citric acid to mustard plants Application of entire dose of EDTA to mustard plants	Doses applied: 0.4 g/kg and 0.8 g/kg of soil Dose applied: 3 g/kg of soil

Table III: Greenhouse Testing Activities (continued)

Date	Activity	Comments
January 10, 2000	Greenhouse visit	Mustard plants that received an EDTA dose were parched
January 12, 2000	Application of second half of both doses of citric acid to the mustard	Doses applied: 0.4 g/kg and 0.8 g/kg of soil
January 21, 2000	Application of first half of both doses of citric acid to corn Application of complete dose of EDTA to corn	Doses applied: 0.4 g/kg and 0.8 g/kg of soil Dose applied: 3 g/kg of soil
January 25, 2000	Greenhouse visit	Corn plants that received an EDTA dose were parched
January 28, 2000	Application of second half of both doses of citric acid to corn	Doses applied: 0.4 g/kg and 0.8 g/kg of soil
February 3, 2000	Harvesting of corn Soil sampling	Only the aerial parts, cut in 5-cm pieces for drying
February 28 to March 2, 2000	All plants pulverized to 2 mm	All samples weighed at this stage
March 16, 2000	Preliminary meeting on drafting of the report	Present: D. Barré, A. Karam, L. Olivier, F. Schooner, N. Moreau and G. Lessard
March 21, 2000	Start of germination tests on both plant species	8 tests in all (4 for mustard and 4 for corn, for each treatment)
April 3, 2000	Harvesting of corn roots (3 samples) Two soil samples taken from the bottoms of the pails (EDTA-corn and EDTA-mustard)	Sampling to determine lead and copper concentrations in the roots of corn plants treated with citric acid or EDTA
April 4, 2000	End of germination tests	All seeds germinated normally and no significant differences between the different conditions were noted

5.2 Fertilization and Soil Amendment

Chemical fertilizers (nitrogen, phosphorus and potassium) were added uniformly to the soil for all samples. Quantities of each of these fertilizers were determined using the recommendations of the Quebec Fertilizer Manufacturers Association for silage corn. The same amounts were used for the mustard.

The recommendation for nitrogen is systematic application of 180 kg/ha, whereas phosphorus and potassium requirements depend on the soil analysis. Table IV shows calculated requirements for the three nutrients.

Table IV: Fertilizer Requirements

Element	Baseline analysis		Recommendation	
	mg/kg	mg/ha*	mg/ha*	mg/kg**
Nitrogen	10	27	180 (N)	66
Phosphorus	42	114	100 (P_2O_5)	37
Potassium	177	480	90 (K_2O)	33

* Calculations based on an average depth of 0.17 m.
** An average density of 1.6 was used to transform volume to weight.

Using the standard depth of 0.17 m as average and an average density of 1.6[2] given the type of soil in the Farnham cell, the following correlation factors were obtained for determining fertilizer requirements:

- 1 kg/ha[3] = 0.368 mg/kg
- 1 mg/kg = 2.720 kg/ha

The fertilizers used (34-0-0 for nitrogen, 0-20-0 for phosphorus and 0-0-50 for potassium) were mixed in the soil together with compost. The total volume of soil used was about 400 L (estimated weight = 640 kg), to which 20 kg (dry weight) of compost were added—30 kg of bark compost with a 35% humidity content. The compost was added to increase the proportion of organic matter in the soil to about 3% (p/p) since the soil was very poor in organic matter.

Based on the recommendations in Table IV, the fertilizers were applied as shown in Table V.

Table V: Quantities of Fertilizer Applied

Type of fertilizer	Amount used* (g)
34-0-0	128
0-20-0	122
0-0-50	44

*For the 660 kg of soil

At the meeting held December 14, 1999, at the Laval University greenhouses, it was decided to make a second application of fertilizer to the corn plants, using the same dose as initially, to increase their biomass. This application was made on January 4, 2000.

2. Density of moderately compacted sand according to W.F. McCollough's Geotechnical Gauge
3. 1 ha: 10 000 m^2 x 0.17 m = 1700 m^3 x 1.6 x 1 000 = 2 720 000 kg

5.3 CITRIC ACID AND EDTA DOSES

Preliminary laboratory tests were performed to check the effect of citric acid on the Farnham soil. The purpose was to determine quantities of citric acid required to lower soil pH enough to increase metal solubility but still remain within the survival range of the plants under study. Another objective was to determine the volume of acidifying solution required to uniformly distribute the liquid without causing excessive leaching beyond the solid matrix.

Given plant survival constraints, target pH was set at 5. The laboratory tests showed that a 0.8 g dose of citric acid was required to lower the pH of 1 kg of soil from 6.1 to 4.93 (Table VI). This was used to establish the double dose of citric acid that would be used during the greenhouse tests. The single dose was then calculated by dividing in two the dose required to achieve a pH of 5.

Last, the volume of solution to add was determined by checking percolation of a liquid through a column of the soil used for the phytoremediation test. Different amounts of water were added to a preset volume of soil and the test amount that yielded the best liquid distribution in the matrix without causing leaching was selected for the greenhouse tests.

Table VI: Citric Acid Dosages

Dose	Amount of Citric Acid (g granular citric acid/kg of soil)	pH
None	0.0	6.1
Simple	0.4	5.35
Double	0.8	4.93

The EDTA dose was set at 3.0 g/kg of soil based on tests with *Brassica juncea* conducted by Ruby Beil (1998). Appendix A shows the results of these tests. All chelates were applied in solution. Table VII shows the doses used.

Table VII : Chelate Dosages

Chelate	Molecular Weight (g)	Dose	Volume of solution added for each test (L)	Molarity[1] of the solution (mol/L)
Citric acid	210.14	Single (0.4 g/kg of soil)	2.5	0.0252
			1.25	0.0504
		Double (0.8 g/kg of soil)	2.5	0.0504
			1.25	0.1008
EDTA	380.18	1 x 3 g/kg of soil	2.5	0.104
			1.25	0.208

1. Molarity: amount of solute per unit volume of solvent, expressed as mols per litre

6 RESULTS

6.1 GENERAL OBSERVATIONS ON PLANT GROWTH

Neither of the species studied showed signs of deficiency or negative reaction to the high concentrations of lead and copper in the soil. Though the soil was very poor in organic matter (less than 3% after application of compost), both species adapted very well to the poor, sandy soil (Appendix B, photos 1 and 2).

Application of EDTA had a lethal effect on the mustard and corn plants (Appendix B, photos 3 and 4). Neither species was negatively affected by the citric acid, however, despite a second application one week after the first.

A growth period of about 50 days (±3 days) for *Brassica juncea* and about 70 days (±4 days) for *Zea mays* was required to obtain a large biomass.

6.2 PHYTOEXTRACTION OF LEAD AND COPPER

Table VIII identifies and briefly describes the 31 plant samples. Tables IX and X show lead and copper contents of the samples.

Table VIII: Sample Identification

Species	Plant part	Sample name	Description
Brassica juncea	Aerial	Control 1	Sample 1/2,* chelate
		Control 2	Sample 2/2, chelate
		EDTA 1	Sample 1/2, one dose of EDTA (3 g/kg of soil)
		EDTA 2	Sample 1/2, one dose of EDTA (3 g/kg of soil)
		Citric acid (X1) 1	Sample 1/2, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X1) 2	Sample 2/2, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X2) 1	Sample 1/2, 2 double doses of citric acid (0.8 g/kg of soil)
		Citric acid (X2) 2	Sample 2/2, 2 double doses of citric acid (0.8 g/kg of soil)
	Root	Control 1	Sample 1/2, no chelate added
		Control 2	Sample 2/2, no chelate added
		EDTA 1	Sample 1/2, one dose of EDTA (3 g/kg of soil)
		EDTA 2	Sample 1/2 one dose of EDTA (3 g/kg of soil)
		Citric acid (X1) 1	Sample 1/2, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X1) 2	Sample 2/2, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X2) 1	Sample 1/2, 2 double doses of citric acid (0.8 g/kg of soil)
		Citric acid (X2) 2	Sample 2/2, 2 double doses of citric acid (0.8 g/kg of soil)
Zea mays	Aerial	Control 1	Sample 1/3, no chelate added
		Control 2	Sample 2/3, no chelate added
		Control 3	Sample 3/3, no chelate added
		EDTA 1	Sample 1/3, one dose of EDTA (3 g/kg of soil)
		EDTA 2	Sample 2/3, one dose of EDTA (3 g/kg of soil)
		EDTA 3	Sample 3/3, one dose of EDTA (3 g/kg of soil)
		Citric acid (X1) 1	Sample 1/3, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X1) 2	Sample 2/3, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X1) 3	Sample 3/3, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X2) 1	Sample 1/3, 2 double doses of citric acid (0.8 g/kg of soil)
		Citric acid (X2) 2	Sample 2/3, 2 double doses of citric acid (0.8 g/kg of soil)
		Citric acid (X2) 3	Sample 3/3, 2 double doses of citric acid (0.8 g/kg of soil)
	Root	EDTA 1	Sample 1/3, one dose of EDTA (3 g/kg of soil)
		Citric acid (X1) 1	Sample 1/3, 2 single doses of citric acid (0.4 g/kg of soil)
		Citric acid (X2) 1	Sample 1/3, 2 double doses of citric acid (0.8 g/kg of soil)

* The numerator indicates the number of the sample and the denominator the number of duplicates.

Table IX : Copper and lead measured in *Brassica juncea*

	Dry weight (g)	Concentration		Total extraction		Performance*	
		Lead mg/kg	Copper mg/kg	Lead (mg)	Copper (mg)	Lead (mg/kg soil)	Copper (mg/kg soil)
Aerial tissue							
Control 1	92.40	20	16	1.85	148	0.06	0.05
Control 2	117.30	16	20	1.88	2.35	0.06	0.07
Control (average)	104.85	18	18	1.86	1.91	0.06	0.06
EDTA 1	84.90	2700	300	229.23	25.47	7.16	0.80
EDTA 2	57.40	2200	260	126.28	14.92	3.95	0.47
EDTA (average)	71.15	2450	280	177.76	20.20	5.55	0.63
Citric acid (X1) 1	117.20	24	16	2.81	1.88	0.09	0.06
Citric acid (X1) 2	123.00	20	20	2.46	2.46	0.08	0.08
Citric acid (X1) (average)	120.10	22	18	2.64	2.17	0.08	0.07
Citric acid (X2) 1	114.10	130	64	14.83	7.30	0.46	0.23
Citric acid (X2) 2	83.80	210	80	17.60	6.70	0.55	0.21
Citric acid (X2) (average)	98.95	170	72	16.22	7.00	0.51	0.22
Roots							
Control 1	11.40	180	16	2.05	0.18	0.06	0.01
Control 2	11.20	170	16	1.90	0.18	0.06	0.01
Control (average)	11.30	175	16	1.98	0.18	0.06	0.01
EDTA 1	10.50	110	28	1.16	0.29	0.04	0.01
EDTA 2	11.00	140	24	1.54	0.26	0.05	0.01
EDTA (average)	10.75	125	26	1.35	0.28	0.04	0.01
Citric acid (X1) 1	13.80	250	20	3.45	0.28	0.11	0.01
Citric acid (X1) 2	12.40	260	20	3.22	0.25	0.10	0.01
Citric acid (X1) (average)	13.10	255	20	3.34	0.26	0.10	0.01
Citric acid (X2) 1	15.00	520	28	7.80	0.42	0.24	0.01
Citric acid (X2) 2	10.30	280	28	2.88	0.29	0.09	0.01
Citric acid (X2) (average)	12.65	400	28	5.34	0.35	0.17	0.01

*Calculated based on the assumption that each 22-L pail holds 32 kg of soil (dry weight); performance was calculated by dividing metal weight (total extraction) by 32.

Table X: Copper and lead measured in *Zea mays*

	Dry weight (g)	Concentration		Total extraction		Performance*	
		Lead mg/kg	Copper mg/kg	Lead (mg)	Copper (mg)	Lead (mg/kg soil)	Copper (mg/kg soil)
Aerial tissue							
Control 1	226.80	40	8	9.07	1.81	0.28	0.06
Control 2	286.70	32	8	9.17	2.29	0.29	0.07
Control 3	252.30	44	8	11.10	2.02	0.35	0.06
Control (average)	255.27	39	8	9.78	2.04	0.31	0.06
EDTA 1	149.70	280	52	41.92	7.78	1.31	0.24
EDTA 2	174.10	600	88	104.46	15.32	3.26	0.48
EDTA 3	130.10	880	92	114.49	11.97	3.58	0.37
EDTA (average)	151.30	587	77	86.95	11.69	2.72	0.37
Citric acid (X1) 1	225.70	68	12	15.35	2.71	0.48	0.08
Citric acid (X1) 2	203.70	52	8	10.59	1.63	0.33	0.05
Citric acid (X1) 3	286.00	32	8	9.15	2.29	0.29	0.07
Citric acid (X1) (average)	238.47	51	9	11.70	2.21	0.37	0.07
Citric acid (X2) 1	212.70	68	12	14.46	2.55	0.45	0.08
Citric acid (X2) 2	246.00	40	8	9.84	1.97	0.31	0.06
Citric acid (X2) 3	267.20	16	8	4.28	2.14	0.13	0.07
Citric acid (X2) (average)	241.97	41	9	9.53	2.22	0.30	0.07
Roots							
EDTA 1	45.8	960	270	43.97	12.37	1.37	0.39
Citric acid (X1) 1	54.2	1600	350	86.72	18.97	2.71	0.59
Citric acid (X2) 1	50.2	2300	610	115.46	30.62	3.61	0.96

* Calculated based on the assumption that each 22-L pail holds 32 kg of soil (dry weight); performance was calculated by dividing metal weight (total extraction) by 32.

7 DISCUSSION

7.1 GENERAL FINDINGS

The initial results of the soil analysis (Table II) showed that lead content had to be lowered by at least 200 mg/kg to meet the decontamination objective for this metal. Lead content of the soil is currently about 1200 mg/kg, whereas criterion C of the MENV policy is 1000 mg/kg in the case of lead. This means that a decontamination performance (amount of contaminant removed per kilogram of soil (dry weight) with each harvest) of about 100 mg/kg of soil would be required to meet the target rehabilitation criterion in a single season, assuming the feasibility of two harvests annually.

The test results (Table IX and Table X) suggest that the best performance was with *Brassica juncea,* using EDTA (3.0 g/kg of soil) as chelating agent. Lead extraction rate with this combination was 5.55 mg/kg of soil. In other words, it would take 36 harvests to reach the objectives set, or 18 years—since two harvests annually appear feasible with mustard.

Analysis of the corn roots show a decontamination performance of 3.61 mg/kg of soil with citric acid (dose = 1.6 g/kg of soil). These results combined with those obtained in the aerial tissue with EDTA suggest the possibility of applying both treatments before harvesting (first citric acid and then EDTA) to obtain maximum performance with each corn harvest. With this method, a performance of 6.33 mg/kg of soil could be obtained by harvesting the entire corn plant (roots and aerial part)—in which case 32 harvests would be required. However, since corn grows more slowly than mustard, only one harvest per year is possible, and this would mean that rehabilitation would take much longer with the corn than with the mustard.

The tests also showed that metals leached out of the upper layers into the lower layers of the soil with the addition of EDTA (see Table II). This was noted with both plant species. This means that chemical decontamination of the Farnham soil with EDTA could be considered if the cost of the chelate is low enough. This is discussed in section 7.2.

The following summarizes the observations that can be drawn from the tests:

- Plants grown in soil treated with chelating agents extracted much more lead and copper than plants grown in the control soil, indicating that chelating agents should be added to the soil.

- Mustard accumulated high concentrations of lead from EDTA-treated soil. In fact, accumulated lead concentrations exceeded minimum lead

accumulation level for plants classified as hyperaccumulators (1000 mg/kg of dry matter), confirming the aptness of Indian mustard as a test plant.

- The plants did not extract enough metal to reduce lead and copper concentrations in the soil after harvesting to an acceptable level (below level C criteria) economically and within a short period of time (see section 7.2).

- The results show that large amounts of chelating agent and numerous growing periods would be required for either of the two plants to extract very large quantities of lead from the soil (upwards of 200 mg lead/kg soil).

- Application of EDTA causes leaching of metals into the lower soil layers. This could be used to decontaminate soil by chemical leaching, if economically feasible.

7.2 COST ASSESSMENT

As mentioned above, phytoremediation does not seem to be an appropriate method for decontaminating the soil in the Farnham cell. A cost assessment was made nonetheless. The most attractive option would be to use Indian mustard, which would yield two annual harvests of about 1.5 tonnes (dry weight of aerial part of the plant) over the 0.2-ha surface of the cell.

The EDTA (at a cost of $8.19/kg) that must be applied to the 2490 tonnes (1556 m^3) of soil to be treated before each harvest at a rate of 3 kg/tonne soil would cost $122 400 a year.

Table XI shows cost estimates for the field operations.

Table XI : Annual Costs with Two Mustard Harvests

Activity	Cost ($C)
Site preparation	800
Fertilizer (nitrogen, phosphorus, potassium)	1 600
Seeding (including seeds)	800
Monitoring (including irrigation)	1 600
EDTA	122 400
Harvest	1 200
Drying	600
Disposal (+ transport)	400
TOTAL	**129 400 $**

*Each cost item includes management.

As outlined above, the annual cost shown in Table XI must be multiplied by 18 to lower the lead content of the soil to meet the level C criterion of the MENV policy. In other words, total cost of phytoremediation of the soil in the Farnham confining cell is clearly prohibitive compared to other options. For example, burying the soil in an appropriate location would cost about $100 000 and would constitute final disposal.

8 RECOMMENDATIONS

The EDTA alone would cost $25 per tonne of soil to be treated, and it must be applied for every harvest—that is, twice a year in the case of mustard. This means that even if it were possible to obtain the EDTA at a better price, phytoremediation, alone, does not seem to be a realistic option for soil remediation at Farnham. The possibility of combining phytoremediation with some other technology (taking advantage of the fact, for example, that the soil is confined in a watertight facility to recover leachate and the solubilized metals it contains) could be considered.

Costs are very high and treatment period long compared with other methods of rehabilitation (secure landfill, for example)—two major factors against phytoremediation compared to the standard technologies that prevent us from recommending it as the sole approach for decontamination of the soil in the Farnham confining cell.

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APPENDIX A

Results obtained by Ruby Beil (1998)

Table 8. Averages and standard deviations of lead in soil, tissue, and leachate and the percent removed from soil and accumulated in the whole plant for each species/EDTA treatment.

Treatment	Initial	Leaf [Pb]	Total	Total	%Removed
Spp./[EDTA]	Soil Pb	at harvest	Plant Pb	Leachate Pb	by whole plant
g EDTA kg^{-1} soil	mg	mg kg^{-1}	mg		%
Mustard—0.0	9748±1671	4.6 ±1.4	1.4± 0.1	0.0±0.0	0.01
Fescue—0.0	10523±463	3.9 ±0.3	3.3± 0.9	0.0±0.0	0.03
Maple—0.0	9823±1383	1.8 ±0.3	0.6± 0.0	0.3±0.4	0.01
Mustard—1.0	9525±730	59.2 ±5.6	6.9± 4.1	0.0±0.0	0.10
Fescue—1.0	9231±2438	131.5 ±5.2	4.4± 0.3	0.0±0.0	0.05
Maple—1.0	10223±625	25.1 ±4.3	8.3± 8.0	16.4±25.1	0.08
Mustard—2.0	10538±198	850.7±235	28.9±10.8	0.0±0.0	0.27
Fescue—2.0	10491±1646	220.4±37.3	59.3± 0.5	0.0±0.0	0.57
Maple—2.0	10333±832	124.1±9.0	6.5± 1.5	9.5± 3.9	0.06
Mustard—3.0	10020±604	1073.5±252	72.0±15.3	0.0±0.0	0.72
Fescue—3.0	9679± 87	315.9±54.3	29.4± 1.8	21.1±55.2	0.30
Maple—3.0	9392±282	78.5 ±68.8	3.2± 1.0	21.4± 9.4	0.23
Mustard—5.0	10457±490	1307.7±174	61.1± 4.7	0.0±0.0	0.59
Fescue—5.0	10351±424	273.4±110	29.2± 9.1	0.0±0.0	0.28
Maple—5.0	10297±1141	90.6 ±56.0	3.0± 0.7	7.4±12.6	0.03
Mustard—8.0	10602±961	1316.2±183	68.2± 5.7	0.0±0.0	0.64
Fescue—8.0	9330±2171	433.6±68.7	35.9± 4.1	0.0±0.0	0.39
Maple—8.0	8973±1422	121.6±52.3	6.0± 1.1	32.0±20.7	0.07
Mustard—12.0	10938±5085	1193.9±138	80.1±10.3	0.0±0.0	0.73
Fescue—12.0	10442±609	402.1 ±26.5	35.9± 1.4	54.1±93.8	0.52
Maple—12.0	10635±2235	175.4 ±145	4.0± 0.4	10.7±3.4	0.10

APPENDIX B

Photographs

Biogenie

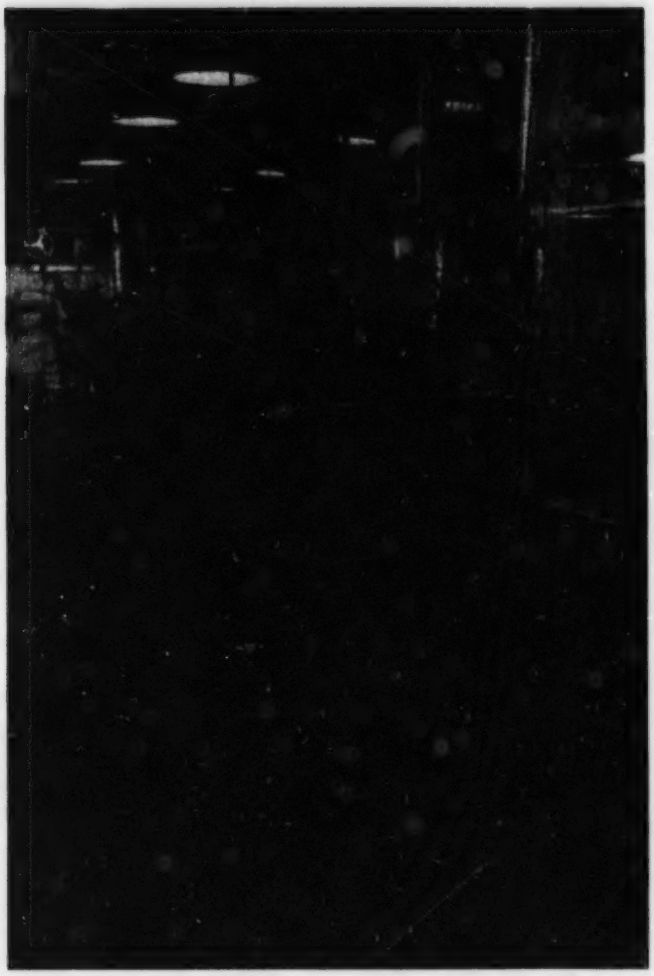

Photo 1: *Brassica juncea* plants at the time of the first chelate application (January 5, 2000).

Biogenie

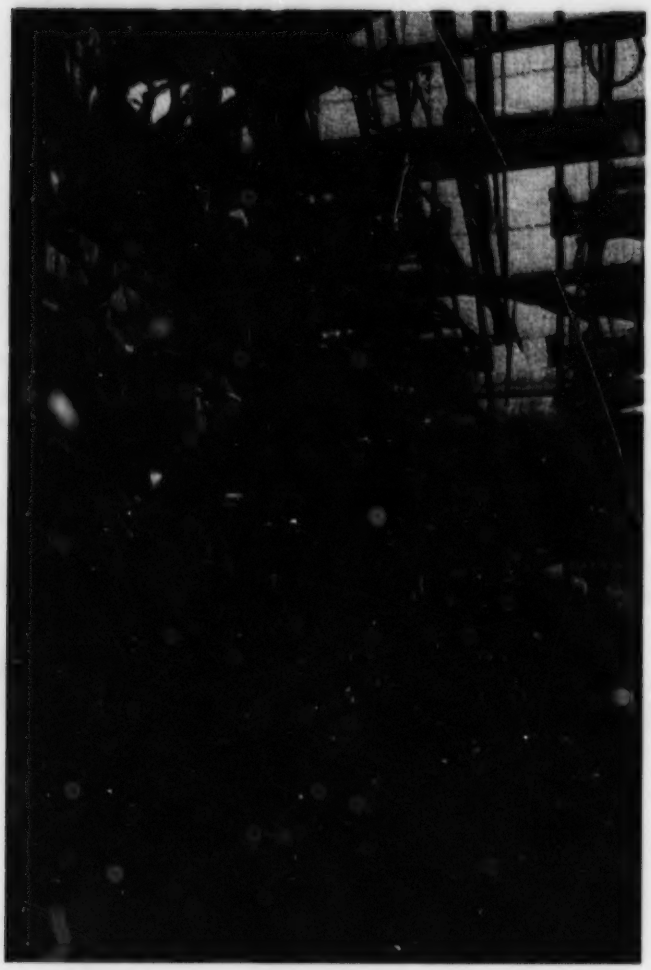

Photo 2: *Zea mays* plants at the time of the first chelate application (January 21, 2000).

Biogenie

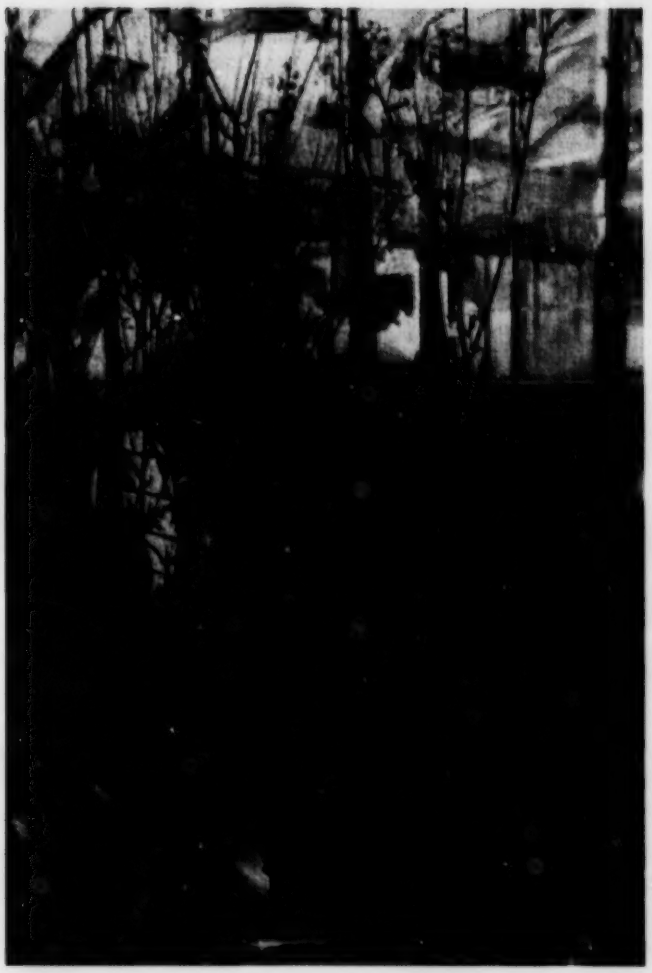

Photo 3: *Brassica juncea* plants five days after application of EDTA (January 10, 2000).

Photo 4: *Zea mays* plants four days after application of EDTA (January 25, 2000).